Oceanic karma? Eco-ethical gaps in African EEE metal cycle may hit back through seafood contamination

Eva Garcia-Vazquez[a1*], Valentin Geslin[a1], Pablo Turrero[b], Noemi Rodriguez[c], Gonzalo Machado-Schiaffino[a], AlbaArdura[a]

a Department of Functional Biology, University of Oviedo, Spain
b Spanish National Distance-Learning University (UNED), Gijón Campus, Spain
c Department of Education Sciences, University of Oviedo, Spain

*Corresponding author egv@uniovi.es
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Abstract
The increasing global demand for electric and electronic equipment (EEE) such as smartphones, tablets and electric car batteries has resulted in an increase in heavy metal releases to the environment at different steps during its manufacture (e.g. mining, extraction, production and e-waste). Some critical raw materials (CRMs) that sup- ply the worldwide demand of technology are mainly sourced from Africa, but their resulting heavy metal pollution can reach citizens from other regions of the world through seafood caught in African waters, which would act as a vector. In this study, we review heavy metal contents in African fish and, as proof of concept, we analyse heavy metal content in three tuna species (*Thunnus alalunga*, *T. albacares* and *T. obesus*) caught in different regions inside the Sustainable Fisheries Partnership Agreements (SFPAs) by Spanish fleets and commercialised in Spain. *Thunnus alalunga* and *T. albacares* from African waters had higher concentrations of heavy metals (especially Hg but also As and Pb) in muscle than samples of the same species caught in other waters. Metal profiles in tunas from African waters were significantly correlated with those of continental and coastal fish from nearby areas impacted by mines and e-waste, as found in the literature review. Based on these results we identify re- search priorities that should be addressed in order to improve the social and environmental sustainability of EEE metal manufacture in Africa.

Key words: Heavy metals, *Thunnus* sp., Fish, E-waste, Metal pollution

1. Introduction

1.1. Metals for electronic products, a circular matter

The European Commission has created a list of critical raw materials (CRMs) for the EU. These materials are needed to produce a broad range of goods used in everyday life and modern technologies, but there is no reliable and unhindered access to them. Four CRM metals from the 2017 EU CRM list are produced mainly in African countries: cobalt in the Democratic Republic of Congo (DRC) and Tanzania, platinum and vanadium in South Africa, and tantalum in the DRC and Rwanda; the CRM list can be found at https://eur-lex.europa.eu/legal-content/EN/TXT/?uri= CELEX:52017DC0490. African countries are also important producers of copper, with copper mines and more than 60% of the world's production of cobalt concentrated in the Copperbelt crossing the south of the DRC and the north of Zambia (Schlüter, 2006; summary data available at https://www.indexmundi.com/minerals/?product=cobalt&graph= production), principally in copper-cobalt ores, while in other zones cobalt is mixed with nickel or arsenic (Roberts and Gunn, 2014).

The demand for these elements increases every year due to their use in electronic devices such as smartphones and in the batteries of electric cars. An increased use of metals like copper and cobalt in electric and electronic equipment (EEE) is significantly correlated with a worsening of sustainable development indicators, copper being the base metal with the broadest impact (combining mined and imported metal) and cobalt the second one (Nansai et al., 2019). Cobalt mining in the DRC illustrates these impacts. Manual extraction and washing of cobalt ore encompass both environmental and human rights threats (Elenge and de Brouwer, 2011; André and Godin, 2014; Amnesty International, 2016), and the further milling and sizing of the ore, and the sulphuric acid leached by slurries during the refining process, increase environmental damages (Roberts and Gunn, 2014). In mining zones, humans are exposed to high concentrations of heavy metals, mainly via water and diet (Banza et al., 2009; Cheyns et al., 2014; Elumalai et al., 2017; Kalonda et al., 2017), which causes severe health problems, especially in children and pregnant women (Squadrone et al., 2016a; Banza et al., 2018; Musa Obadia et al., 2018). Besides representing a serious risk for human health, mining has also been suggested as a cause of fish declines in African rivers through waterborne metal exposure that endangers fish populations (e.g. Kambole, 2003; Sracek et al., 2012; Jackson et al., 2016).

Metals extracted in Africa, like cobalt and tantalum, follow a route that involves big Asiatic companies as intermediaries (e.g. Gulley et al., 2019; Shütte, 2019). These buy the minerals and produce the components of batteries and other EEE, and these are in turn sold to companies in the electronics, electrical and transport sectors. These companies then sell the final product (smartphones, tablets, electric cars) worldwide, with many consumers living in North America and in European countries. The journey of EEE minerals that starts in African mines ends with the export of that EEE, once it becomes obsolete (called generically e-waste), back to the African continent. This is important for the economies of recipient countries, but at the same time represents an environmental burden in areas where recycling is not sufficiently developed. As an example, Ghana has one of the biggest e-waste landfills in the world, which receives mainly European electronic waste. Kyere et al. (2016, 2017) demonstrated that Ghana's e-waste deposits cause significant soil pollution by heavy metals, and Cao et al. (2020) found a significant hazard index (i.e., health risk) due to Cu, As, Cd, Sb and Pb bioaccessibility in e-waste burning sites at Agbogbloshie (Accra, Ghana); however, local populations are largely unaware of the environmental problems posed by such e-waste (Owusu et al., 2017). The disposal of European e-waste in African countries closes a circle that is an example of “not in my backyard” global policy. The enormous environmental toll of EEE minerals is paid by producer and e-waste recipient countries in Africa, while European countries benefit from new devices and green no-carbon transport thanks to electric cars that carry African CRM in their batteries.

1.2. Potential drawbacks of the “not in my backyard” policy

Perhaps the circle is not perfect and EEE metals can reach European citizens: seafood could be a possible vector of heavy metal pollution between Africa and Europe. Heavy metals produced in mining and e-waste areas arrive in coastal waters through rivers and river plumes. Since these metals are less bioavailable in saltwater than in freshwater, biota exposed to marine sediments is the main concern regarding bioaccumulation (e.g. Ansari et al., 2004). Although not directly exposed to sediments like benthic organisms, pelagic species can also incorporate metal pollutants in their tissues through the trophic chain. Metals start accumulating in the first food-web links (i.e., plankton: e.g. Srichandan et al., 2016), then reach higher trophic levels like fish (Le Croizier et al., 2016), and can end in top predators; metal accumulation then depends on species (for instance, because of their trophic level and life history, including foraging habits: e.g. Le Croizier et al., 2016, 2019). An example of highly predatory fish that accumulates heavy metals is tuna, whose mercury content can be dangerous for consumers (e.g. Licata et al., 2005; Araújo and Cedeño-Macias, 2016; Cammilleri et al., 2018). Metal accumulation in tuna depends on the species, each exhibiting specific patterns; for example, Atlantic *Thunnus obesus* has more Hg but less Cu than *T. alalunga* (Besada et al., 2006), while in northwestern Spain T. alalunga exhibited higher contents of both As and Pb than *T. albacares* (García et al., 2016). Differences between sexes in metal levels within a tuna species (*Thunnus thynnus*) have been also reported (Di Bella et al., 2015). On the other hand, there are regional differences in the level of heavy metals within a species (e.g., in *T. albacares*: Nicklisch et al., 2017), supporting the idea that metal intake depends, among other factors, on different environmental levels due to anthropogenic activities (Lamborg et al., 2014).

Fish caught in marine African waters enter the global trade and can be sold anywhere, but are likely to turn up in Europe. EU Sustainable Fisheries Partnership Agreements (SFPAs) allow EU vessels to catch tuna (*Thunnus* sp.) as they migrate along African waters. The Food and Agriculture Organization of the United Nations (FAO) divides the world's oceans into several “major fishing areas for statistical purposes” (http://www.fao.org/fishery/area/search/en). SFPAs in West African waters include tuna agreements with Cape Verde, Senegal, Gambia (these also allow to catch hake), Liberia and Ivory Coast within Fishing Area 34; and Sao Tomé e Principe in the northern zone of Fishing Area 47. Multispecies agreements to allow the catch of a variety of fish stocks (including tuna) in the country's exclusive economic zone are established with Morocco, Mauritania and Guinea Bissau, all within Fishing Area 34 (https://ec.europa.eu/fisheries/cfp/international/ agreements_en).

If African heavy metal pollution reaches tuna in the open seas through river plumes and the trophic chain, Europe could be suffering EEE metal pollution through the exploitation of marine species from these SFPAs. This could be investigated by analysing metals in tuna (and hake) caught from African SFPA zones and comparing their profiles with those of tuna caught in other areas, and with fish from African continental waters exposed to EEE metals. This approach should take into account that metal content may differ between tuna species (e.g. Besada et al., 2006; Nuñez et al., 2018), thus considering tunas separately. To explore this idea with a proof of concept, we carried out a literature review of heavy metal content in African fish and analysed heavy metals in tuna caught by Spanish fleets in different SFPA regions and commercialised in Spain, to test if the main pollutants of African fish were also more concentrated in pelagic tunas.

2. Material and methods

2.1. Review of heavy metal pollution in African fish

A systematic search was conducted in the database Google Scholar between January 2020 and March 2020. The terms used in the query were “metal”, “contamination”, “fish” and the name of the country combined with “AND”. After an initial screening, all potentially eligible articles were downloaded. Data in the selected articles were extracted and compiled in a table including the following columns: country name; location, latitude and longitude of sampling site; year of sampling; class, order, family, name and FAO group of sampled species; average concentrations of selected metals (As, Cd, Co, Cr, Cu, Hg, Ni, Pb, Zn); standard deviations, minimum values and maximum values of those concentrations; units expressed either in dry weight (DW) or wet weight (WW); and reference of the article. Maps showing heavy metal concentrations were constructed using the software R (R Core Team, 2018) and the package ggmap (Kahle and Wickham, 2013).

The analysis of the metadata gathered in the previous step started by classifying the sources of pollution identified by the authors of each article. The stated sources were very diverse, because our data were gathered on a continental scale, thus sources were classified according to three categories: mining (when resource extraction or mining- associated processes were explicitly mentioned in the article as a source of pollution), industrial (including industry and shipping-ship construction as associated processes; explicitly identified in the article as pollution sources in the sampling area) or diffuse pollution (agriculture and urban waste runoff, geological background; stated in the article as pollution sources).

2.2. Sampling

A total of 52 true tuna samples (genus *Thunnus*) fished by Spanish vessels and commercialised in Spain were randomly obtained from local supermarkets in Asturias, northern Spain, between January and March 2019; Table 1 includes all the relevant information contained in their labels.

Although *Thunnus* species are highly migratory, the three species re- main at least for some time in African catch areas where they are exposed to metal pollution through the food chain: the main spawning area for *T. albacares* in African waters is in the Gulf of Guinea, where they concentrate after their long migrations to and from America when they are around three years old (Zagaglia et al., 2004); immature *T. alalunga* grow in front of the Moroccan and Mauritanian coasts be- tween 22° and 32° latitude N, where they are fished before moving to their spawning areas (Nikolic et al., 2017); and population genetic data suggest that *T. obesus* also have a spawning and nursery area around the Gulf of Guinea (Gonzalez et al., 2008).

2.3. DNA extraction and barcoding

All samples were analysed for species authentication. DNA extraction was performed following the protocol for Chelex® resin (Bio-Rad Laboratories) developed by Estoup et al. (1996). A fragment of control region gene was amplified by polymerase chain reaction (PCR), employing Viñas and Tudela (2009)'s primers L15998 (5′-TACCCCAAA CTCCCAAAGCTA-3′) and CSBDH (5′-TGAATTAGGAACCAGATGCCA G- 3′). PCR conditions were the following: an initial denaturing step of 5 min at 94 °C, followed by 35 cycles of denaturing at 94 °C for 45 s, annealing at 50 °C for 45 s and extension at 72 °C for 1 min. and a final ex- tension at 72 °C for 5 min. All the PCR assays were run in a Thermal Cycler (Applied Biosystems, model 2720).

PCR products were separated and visualized using 1.5% agarose gel stained with 10 mg mL^{-1} SympleSafeTM (2.5 µL, EURx, Gdansk, Poland). Amplicons were sent for sequencing at Macrogen Spain, Inc. (Madrid, Spain) using a standard Sanger sequencing method procedure.

Sequences were edited and trimmed employing the BioEdit Sequence Alignment Editor software (Hall, 1999) and aligned with the ClustalW application included in BioEdit. The outcome sequences were identified in the GenBank database using the BLAST algorithm (http://www.ncbi.nlm.nih.gov/genbank/).

2.4. Heavy metal content analysis

From each sample, 0.5 mg were digested with a mixture of HNO_3 (7 mL) and H_2O_2 (1 mL) in a microwave digestion system (Milestone HPR-FO-20) at 15000 W for 0.5 h. The concentrations of several metals (As, Cd, Co, Cr, Cu, Hg, Ni, Pb, Zn) were measured using inductively coupled plasma mass spectrometry in Agilent 7700 Series ICP-MS.

The sensitivity of this method was determined according to the detection limits established for this spectrometer, which were 0.0124 µg/kg for Cr, 0.009031 µg/kg for Co, 0.01331 µg/kg for Ni, 0.01454 µg/kg for Cu, 0.08849 µg/kg for Zn, 0.04129 µg/kg for As, 0.01523 µg/kg for Cd, 0.02804 µg/kg for Hg and 0.007041 µg/kg for Pb.

2.5. Quality assurance and quality control

Quality assurance and control of heavy metal analyses was carried out as in Gu and Gao (2018). Reagent blanks were carried out in every analysis and applied to correct the analytical results whenever required. Analysis of five metals in fish muscle was performed on the certified European Reference Material ERM® BB422 (which is fish muscle from saithe Pollachius virens) as well as on samples for quality control.

The muscle samples were analysed in triplicate as in Wang et al. (2020). To assess the method's precision, the coefficient of variation (CV) of the average of three replicates of a sample solution was determined. The calculated CV was different for every element and species analysed (see 3.2).

Comparisons of measurement results with the certified values were done following the European Reference Materials (ERM) guidelines. Briefly, to evaluate method performance, the absolute difference be- tween the mean measured value and the certified value (Δm) is com- pared with the expanded uncertainty of difference between result and certified value ($U\Delta$), corresponding to a confidence level of approximately 95%. If $\Delta m \leq U\Delta$ then there is no significant difference between the measured result and the certified value. The results were in good agreement with the certified value for all elements, except for mercury (see Table 2). The ratio [measured concentration / certified concentration] was 1.28 for this metal.

2.6. Maximum tolerable weekly intake calculation

The Maximum tolerable Weekly Intake (MWI), as average was calculated following the Joint FAO/WHO Expert Committee on Food Additives and Contaminants (2011) for an adult of 70 kg using the formula:

$$MWI = (PTWI \times BW) / MHM$$

where PTWI is the Provisional Tolerable Weekly Intake (5 μg/kg of total Hg, 25 μg/kg of Pb, 6–8 μg/kg of Cd), BW is Body Weight, and MHM is the Median concentration of the Heavy Metal in the seafood product.

2.7. Statistics

In the metadata analysis, comparisons of the proportions of safe and unsafe mean metal contents between groups of samples were done employing contingency Chi square methodology, using the standards set in the European legislation (maximum levels allowed in fish considered fit for consumption: 0.1 mg/kg of Cd, 0.5–1 mg/kg of total Hg and 0.3 mg/kg of Pb; EC No 1881/2006, available at https://eur-lex.europa. eu/legal-content/ES/TXT/HTML/?uri=CELEX:32006R1881&from=ES, accessed June 2020).

Comparisons between group distributions for categorical variables were also performed using contingency Chi square tests. The best correlations between the heavy metal profiles of different samples or groups of samples were chosen based on Akaike Information Criteria for linear model fit, and r or rank ϱ coefficients were calculated. The conventional threshold of $p = .05$ was considered to indicate statistical significance.

In tuna samples, a global comparison of the six groups of samples for all or various metal contents was done using a MANOVA analysis with Wilk's lambda. For post-hoc pairwise comparisons between groups of samples we used Mahalanobis distances. The analysis of different metals in T. alalunga and T. albacares was performed through two-way ANOVA, with factors ‘species’ and ‘continent’ (two levels: African versus non African).

A Principal Component Analysis (PCA), using the correlation option and mean value imputation for missing values, was carried out to visualize the global variation of the dataset. Kaiser–Meyer–Olkin (KMO) and Bartlett's sphericity tests were first conducted to assess the validity of PCA (as in Gu et al., 2016). All these analyses were performed with the PAST software (Hammer et al., 2001).

3. Results and discussion

3.1. Heavy metal pollution in African fish

A meta-analysis of 689 data entries from 83 publications (the raw database can be found in Geslin and Garcia-Vazquez, 2020; original data were transformed to mg/kg) revealed that most studies focused on cadmium, copper and lead, and were done on the following countries: Algeria, Cameroon, DR Congo, Egypt, Ethiopia, France (Reunion is- land), Ghana, Ivory Coast, Jordan, Kenya, Mauritania, Mauritius, Morocco, Mozambique, Nigeria, Oman, Saudi Arabia, Senegal, South Africa, Tanzania, Togo, Tunisia and Yemen. Data for arsenic and cobalt, which reach high concentrations in some areas, were less numerous than data for other metals (Fig. 1A). Most polluted fish were concentrated in zones rich in mines (DRC Congo, Guinea, Sierra Leone, Tanzania; Fig. 1B) and near river plumes (the Casamance, Congo, Niger, Nile and Zambezi rivers, the Senegal-Gambia catchment area). Accumulation of copper and cobalt was found principally, but not only, in fish caught from land waters inside the Copperbelt and in Ghana, an important e-waste importer. This illustrates and indirectly supports the idea of EEE metals as drivers of pollution, and would sug- gest river plumes as ways for heavy metals to travel to the sea. The rough profile of heavy metals of fish from Moroccan and Mauritanian waters would be Cu≈Hg≈As>Cd>Pb>Co,while in the Gulf of Guinea the profile would be As>Hg≈Cu>Pb>Co>Cd, in Tanzania it would be Pb>Cu≈As≈Cd>Hg≈Co, and so on (Fig. 1). From the Gulf of Guinea southwards there is a large gap in the geographical coverage of fish metal contents.

In Africa and elsewhere, soil and water pollution by the six metals reported in Fig. 1 have been associated to mining and e-waste (e.g. Kyere et al., 2016, Gbogbo et al., 2017, Kalonda et al., 2017, Ackah, 2019), and fish pollution is the logical consequence of exposure to them. However, pollution close to a mine is not necessarily a result of the extraction process and may not necessarily be caused by the extracted resource: it can also be the product of ore treatments or further metal processing. For example, mercury is associated to gold mining be- cause it is employed in the separation of gold from the ore (e.g. Esdaile and Chalker, 2018). Furthermore, there are other sources of metal pollution in Africa, such as insufficiently controlled industrial development (e.g. Orisakwe, 2014), urban and domestic wastes (e.g. Ouali et al., 2018), agricultural practices (e.g. Ekeanyanwu et al., 2015) and the geo- logical background (e.g. Biney et al., 1994). It is difficult to generalise the contribution of mining to pollution, and other sources should be considered.

An analysis of the meta-dataset grouped by decades and pollution source revealed that the majority of samples from industrial areas (46% of all the samples) were analysed in the last two decades, while the study of pollution caused by mines (24.5% of samples) was more in- tense in the 90s and 2000s and studies on diffuse pollution (29.5% of samples) were carried out principally in the last decade (Fig. 2A). As commented above, not all the studies considered the same metals, and the number of samples analysed for each metal over the studied period was different (As: 67 samples, Cd: 302, Co: 41, Cr: 159, Cu: 255, Hg: 154, Ni: 91, Pb: 368, Zn: 270). The metal profile of the groups of samples subjected to diffuse, industrial and mining pollution was not the same (Fig. 2B): higher Pb and Cd levels characterized pollution associated to mining, Cu and Co levels were higher under industrial pollution, and Cr levels were greater under diffuse pollution (Fig. 2B).

Considering the three metals with legal limits in fish flesh in European regulations, the percentage of samples whose average con- tent was above safe limits under diffuse, industrial and mining pollution was respectively 34%, 20% and 50% for Cd; 32%, 25% and 20% for Hg; and 30%, 23% and 47% for Pb.

3.2. DNA and heavy metal analyses in commercial tuna

The three tuna species were unequivocally identified from DNA sequences and BLAST. The sequences are available in GenBank with accession numbers MT711122- MT711161. Species names stated on the labels were all confirmed genetically, the best hit exhibiting at least 99% identity with the problem sequence and highly significant score. Anecdotally, the bigeye tunas were labeled as “Thunnus obsesus” (Thunnus obesus).

Metal content averages in the five groups of samples (organized by species and capture area; see Table 3) were significantly different (MANOVA with Wilk's $\lambda = 0.041$, $F(36,147.9) = 5.51$, $p = 5.1*10^{-11}$), as expected because the groups are from different species and regions. In T. alalunga and T. albacares, individuals caught in African waters exhibited a higher content of both Hg and Pb than their conspecifics caught elsewhere (North Azores or Cook Islands; see Fig. 3). Focusing only on the three metals that are routinely surveyed as food contaminants because of their high toxicity (Cd, Hg and Pb), differences among groups were also highly significant (Wilk's $\lambda = 0.318$, $F(12,119.4) = 5.4$, $p = 2.9*10^{-7}$). Post-hoc pairwise tests between African and non-African samples of the same species for the contents of the three metals were significant, squared Mahalanobis distances being $D_M = 6.01$ with $p = .026$ for T. alalunga and $D_M = 7.18$ with $p = .017$ for T. albacares.

A two-factor ANOVA of T. alalunga and T. albacares samples (Table 4) showed significant differences by continent for Cu (lower content in African tunas), Hg and Pb (higher content in African tunas, Table 3), and by species for As, Cr, Cu, Hg and Zn. Higher Pb contents were found in African samples than in tuna from other waters, with no significant differences between species (Table 4). T. alalunga had more arsenic than the other two species (Table 3). A significant interaction between factors was found for As (Table 4), for which African samples of T. alalunga and non-African samples of T. albacares were respectively the most polluted within species (Table 3). Remarkably, despite the small FAO 47 sample size, its mean As content was significantly higher than that of the North Azores sample ($t = 3.4$, 15 d.f., $p = .004$). Mean Hg content was significantly higher in African than in other samples, and in T. albacares than in T. alalunga (Table 4). These results confirmed the expected differences between species and a consistent trend of higher Hg and Pb contents in specimens caught in African (versus non-African) waters.

Sixty percent of Cd, 23% of Pb and 84.6% of Hg values obtained for African T. albacares were above the maximum value found by Galimberti et al. (2016) in samples of this species imported to the EU, which were in average 8 µg/kg of wet weight (range 5–13) for Cd, 10 (0–47) for Pb and 350 (254–430) for Hg. In contrast, only 42% of the samples from the Cook Islands exceeded that maximum for Hg. Our Hg data for FAO 34 were higher than those found by Nicklisch et al. (2017) in T. albacares from north-western (total Hg mean of 0.206 ± 0.035 mg/kg) and south-western (0.348 ± 0.101 mg/kg) African wa- ters, and also higher than those found by in T. albacares in Lakshadweep, India (Dhaneesh et al., 2014); and were comparable to the results Kojadinovic et al. (2007) obtained from samples from Mozambican and Reunion Island waters.

The results for T. alalunga in our study were also comparable with those of other authors: Nuñez et al. (2018) found a mean of 1.28 mg/kg of As, 0.014 of Cd and 0.013 of Pb in T. alalunga products sold in Spanish markets, and García et al. (2016) found 0.332 mg/kg of Hg in the same products; these values are similar to the averages found in our study in the Azores samples.

Regarding T. obesus, a species listed as vulnerable by the IUCN in 2011 (Collette et al., 2011), the few samples analysed in this study (from FAO fishing area 34) exhibited the highest Cd and Hg contents and the lowest Pb content of the three species (Fig. 3), which is consistent with interspecific differences in metal content in tuna (e.g. Besada et al., 2006; Nuñez et al., 2018 and references therein). The results for As, Cd, Cu and Zn were within the ranges reported by Chen et al. (2018) for T. obesus caught in the same zone, and generally higher Hg values than those obtained by Torres et al. (2016) in samples from the Azores area and by Sika et al. (2014) on samples caught in Ivory Coast waters between 2011 and 2013, perhaps indicating increasing anthropogenic Hg in African waters. However this has to be taken with caution given the small size of our T. obesus sample.

3.3. Spatial differences in metal content profiles

KMO and Bartlett's analysis indicated that PCA was applicable to the dataset (KMO: 0.70, acceptable; Bartlett's sphericity test = 2435.8 with $p \ll 0.001$). To visualize the relative contribution of the six most studied metals in African waters (Fig. 1) to the variation in the contents of analysed tuna we constructed a scatter plot of Principal Components PC1 and PC2 (Fig. 3), which explained 40.1% and 26.9% of the total variance, respectively. A complete separation between the African tuna and the rest of the samples analysed here was not expected due to the highly migratory behaviour of tunas (T. albacares even undertakes transatlantic displacements, e.g. Maury et al., 2001), but the three species spend significant time in African waters (e.g. Zagaglia et al., 2004; Gonzalez et al., 2008; Nikolic et al., 2017), and the plot points at a somewhat different pattern of metals in tuna from West African waters.

The relationship between the pollution profiles of tunas in this study (as mean content in muscle) and the profiles of African fish from areas near the zones where tunas were caught was explored employing rank correlations. For this, tuna mean contents of the six metals in Fig. 1 were transformed into a rank scale from 1 to 6 for each group of samples and compared with the profiles for fish from in Morocco-Mauritania (FAO 34: Cu≈Hg≈As>Cd>Pb>Co) and the Gulf of Guinea (southern FAO 34 and northern FAO 47: As>Hg≈Cu>Pb>Co>Cd). The non-parametric rank coefficient correlation between the profiles from Morocco-Mauritania and from the Gulf of Guinea was not significant ($\varrho = 0.61$, $p = .08$). The profile of both tuna species caught in FAO 34 was significantly correlated with that of Moroccan-Mauritanian fish ($\varrho=0.89$, $p=.01$ and $\varrho=0.75$, $p=.03$ for T.albacares and T.obesus. respectively), but not with the profile of fish from the Gulf of Guinea ($\varrho = 0.55$ and $\varrho = 0.24$ respectively). T. alalunga caught in the northern FAO 47 exhibited a significant correlation with the Gulf of Guinea profile ($\varrho = 0.83$, $p = .02$), and also, although weaker, with the Morocco- Mauritania profile ($\varrho = 0.75$, $p = .03$). Of course, correlation does not imply causation, and there are no similar environmental causes that could be invoked to explain the correlated pollution profiles. However, these significant positive correlations suggest, and do not discard, the possibility of continental pollution influencing the pollution profiles not only of the fish living there but also those found in tuna caught in nearby oceanic waters.

3.4. Fingerprint analysis of pollution source

According to the geographical concentration of mines shown in Fig. 1B, mining pollution should affect the waters in the Gulf of Guinea more than the Morocco-Mauritania region, which has fewer mines. In order to examine this, we performed a fingerprint analysis (as in Wang et al., 2020) in our tuna samples (using corrected values) and compared it with the heavy metal fingerprints of average African fish subjected to mainly diffuse, industrial or mining pollution, as seen in Fig. 2B and discussed in 3.1. The heavy metal fingerprints of FAO 34 T. albacares were similar to those of fish from locations with a dominantly diffuse pollution profile (Fig. 4). For this T. albacares sample the highest correlation was obtained with the diffuse pollution profile ($r = 0.96$, $p \ll 0.001$), and it had the best AIC of the three possibilities (AIC = 62.5 versus 349.5 and 244.6 for linear correlations with industrial and mining pollution profiles respectively). The same was found for T. obesus from FAO 34, as seen in Fig. 4 ($r = 0.95$, $p \ll 0.001$, AIC = 81.03 with diffuse pollution, versus AIC = 368.5 and 260.8 for the respective correlations with industrial and mining pollution). T. alalunga from FAO 47 had the best-fit correlation with mining pollution ($r = 0.98$ with $p \ll 0.001$ and AIC = 54.2 versus 59.5 for the correlation with diffuse pollution and 229.3 for that with industrial pollution), as also reflected in the fingerprint analysis (Fig. 4).

All together, these results point at higher Hg and Pb pollution in tunas fished from African waters than in other regions, and the metal profiles of African tuna were significantly correlated with the profiles of African fish reported from continental and nearby coastal areas, as well as with the profile reported for African fish subjected to the main pollution source in those areas. Anthropogenic Hg is increasing in all the oceans (Lamborg et al., 2014), and in Africa it could increase due to recognised mercury sources such as industrial waste and shipping, as happens in Tunisia and Algeria (Zohra and Habib, 2016; Bachouche et al., 2017). In the case of Pb, mining could be a large contributor (see Fig. 2 and Section 3.1), and large marine pelagic fish from coastal waters could incorporate lead through the food chain, as reported in many oceanic regions and also in African waters (e.g. Kojadinovic et al., 2007; Le Croizier et al., 2016, 2019).

3.5. Implications for consumers

EU regulation of contaminant levels in food products allows maxi- mum concentrations of 0.1 mg/kg of Cd, 1 mg/kg of total Hg and 0.3 mg/kg of Pb in marine fish, including Thunnus spp. Using uncorrected Hg values, four tunas from FAO 34 were above those limits for Hg: two T. albacares (1.21 and 1.03 mg/kg) and two T. obesus (1.33 and 1.27 mg/kg), and none from other waters. With corrected Hg values one T. obesus remained above the threshold (1.04 mg/kg) and another one was at the limit (1 mg/kg), as was one T. albacares specimen (0.95 mg/kg), while the other remained below the threshold (0.8 mg/kg). One T. alalunga from FAO 27 was above the permitted Cd concentration (0.158 mg/kg).

The Maximum tolerable Weekly Intake for an adult of 70 kg varied depending on the specific metal (Table 5). Hg was the most limiting element, while Pb did not limit consumption recommendations. Following estimations based on uncorrected Hg median values, MWI varied from 276 g of T. obesus from FAO 34 (roughly one big serving size) to more than 1 kg and 600 g of T. alalunga from the Azores. Using corrected Hg values, the MWI for T. obesus from FAO 34 was 353 g, which would correspond to one and a half servings per week. As reported by other authors, most samples of tuna sold in Europe were within safe limits for consumption (e.g. García et al., 2016, Galimberti et al., 2016, Nuñez et al., 2018) and only a few surpassed the allowed limits of metal concentrations. However, some precautions should be taken with the total consumption of tuna and of T. obesus in particular, since it can exhibit high concentrations of different heavy metals (e.g. Torres et al., 2016; Chen et al., 2018). Furthermore, since this species is in a vulnerable conservation status, its fisheries should be strictly controlled.

3.6. Research gaps and current challenges

Research gaps are of a multidisciplinary nature. In the field of environmental pollution, the main knowledge gap is the lack of understanding of the real impact of EEE metal mining and e-waste on fish species. On one hand, only a few metals have been studied (Cd, Hg and/or Pb, e.g. Galimberti et al., 2016, Musa Obadia et al., 2018). Studies should expand to other metals such as As, Co, Cu, Zn and others that can reach toxic levels near mines and e-waste landfills (Banza et al., 2009; Kyere et al., 2016; Ackah, 2019). On the other hand, the geographical coverage of the studies on fish metal pollution in Africa is irregular and has large gaps in many regions; moreover, studies of mining-polluted areas are scarce in comparison with other types of pollution, and so specific effort should be made to cover lakes, drainage systems, basins and coasts potentially affected by mining activity.

With a focus on the possible transfer of EEE metals and e-waste pollution to marine species, studies generally consider only edible species of commercial value. Other marine fish, shellfish and algae, i.e. species occupying different trophic niches should be analysed in order to have an ecosystem-based approach. River, estuarine and marine water and sediments should be studied at the same time. For highly migratory species like tunas and hakes, feeding habits and migration behaviour should be considered; those feeding on prey that grows close to coasts and river plumes will likely be the most affected by EEE metal pollution via the trophic chain.

Regarding environmental health, the risk of metal ingestion through seafood should be investigated in producer and importer countries. Re- search should focus on metal accumulation in edible parts, such as fish muscle and the harvested parts of algae. Mining and e-waste are complex sources of pollution where different metals are involved, thus attention should be paid to mixtures and additive effects between metals (Wang et al., 2005; Saha and Zaman, 2013). In Europe some assessments of heavy metal exposure risk are based on national statistics of fish consumption (e.g. Squadrone et al., 2016b); however, fish con- sumption may vary greatly among locations in a country and among population sectors (e.g. Von Stackelberg et al., 2017). Risk should then be modelled from surveys across different population sectors, and a gender perspective should be adopted, because pregnancy is a stage particularly sensitive to heavy metal exposure (Gull et al., 2018), and the problem is hitting African countries especially hard (Anyanwu et al., 2018).

Research on economy, sociology and politics in producer countries is needed to ensure that promoting cleaner EEE metals does not harm the poorest families. As an example, the transparent inclusion of artisanal cobalt in electric vehicle supply chains is posited to improve the bargaining power of (and, subsequently, the conditions in) local com- munities in the DRC (Zeuner, 2018).

Last but not least, countries should be legally bound to an eco-ethical disposal of their e-waste, to prompt the recycling of electronic components where e-waste is produced instead of exporting them to third countries. In Europe, the circular economy of electronic devices has been recently considered a priority (European Commission, 2020), and countries should implement e-waste policies accordingly.

4. Conclusions

Metal pollution caused by deficient environmental management in mines and e-waste may arrive to oceanic waters and accumulate in fish exported to Europe, as suggested from a proof of concept analysis of tuna commercialised in Spain. Tuna from African waters exhibited higher concentrations of metals in muscle, especially Hg and Pb, than samples of the same species caught in other waters; their metal profiles were significantly correlated with those reported in continental and coastal fish near tuna capture zones, and in African fish exposed to de- fined pollution sources in nearby areas. Research priorities identified here include: 1) to improve the geographical coverage and increase the number of metals analysed in African fish, with a focus on mining regions; 2) to understand the transfer of river and land pollution to sea- water and its accumulation in the marine trophic chain; 3) to assess the risk of heavy metal ingestion derived from the consumption of sea- food polluted with e-waste metals; and 4) to investigate how to improve the social and environmental sustainability of mining and e-waste treatment in producer and importer countries.

CRediT authorship contribution statement

Eva Garcia-Vazquez: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Supervision, Writing - original draft. Valentin Geslin: Data curation, Formal analysis, Investigation, Methodology, Visualization, Writing - review & editing. Pablo Turrero: Conceptualization, Writing - review & editing. Noemi Rodriguez: Investigation, Writing - review & editing. Gonzalo Machado-Schiaffino: Funding acquisition, Writing - review & editing. Alba Ardura: Funding acquisition, Investigation, Methodology, Supervision, Visualization, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

Ackah, M., 2019. Soil elemental concentrations, geoaccumulation index, non- carcino- genic and carcinogenic risks in functional areas of an informal e-waste recycling area in Accra, Ghana. Chemosphere 235, 908–917.

Amnesty International, 2016. “This Is What We Die for”. Human Right Abuses in the Dem- ocratic Republic of the Congo Power the Global Trade of Cobalt. Amnesty Interna- tional ©, Index: AFR 62/3183/2016, London.

André, G., Godin, M., 2014. Child labour, agency and family dynamics: the case of mining in Katanga (DRC). Childhood 21, 161–174.

Ansari, T.M., Marr, I.L., Tariq, N., 2004. Heavy metals in marine pollution perspective–a mini review. J. Appl. Sci. 4, 1–20.

Anyanwu, BO, Ezejiofor, AN, Igweze, ZN, Orisakwe, OE, 2018. Heavy metal mixture expo- sure and effects in developing nations: an update. Toxics 6 (4), 65. https://doi.org/ 10.3390/toxics6040065.

Araújo, CVM, Cedeño-Macias, LA, 2016. Heavy metals in yellowfin tuna (Thunnus albacares) and common dolphinfish (Coryphaena hippurus) landed on the Ecuadorian coast. Sci. Total Environ. 541, 149–154.

Bachouche, S., Houma, F., Gomiero, A., Rabah, B., 2017. Distribution and environmental risk assessment of heavy metal in surface sediments and red mullet (Mullus barbatus) from Algiers and BouIsmail Bay (Algeria). Environ. Model. Assess. 22, 473–490. https://doi.org/10.1007/s10666-017-9550-x.

Banza, C.L.N., Nawrot, T.S., Haufroid, V., Decrée, S., De Putter, T., Smolders, E., Kabyla, B.I., Numbi, O.L., Ilunga, A.N., Mutombo, A.M., Nemery, B., 2009. High human exposure to cobalt and other metals in Katanga, a mining area of the Democratic Republic of Congo. Environ. Res. 109, 745–752.

Banza, C.L.N., Casas, L., Haufroid, V., De Putter, T., Saenen, N.D., Kayembe-Kitenge, T., Musa Obadia, P., Mukoma, D.K.W., Ilunga, J.M.L., Nawrot, T.S., Numbi, O.L., Smolders, E., Nemery, B., 2018. Sustainability of artisanal mining of cobalt in DR Congo. Nat. Sus- tain. 1 (9), 495–504.

Besada, V., Gonzalez, J.J., Shultze, F., 2006. Mercury, cadmium, lead, arsenic, copper and zinc concentrations in albacore, yellowfin tuna and bigeye tuna from the Atlantic Ocean. Cienc. Mar. 32, 439–445. https://doi.org/10.7773/cm.v32i22.1083.

Biney, C., Amuzu, A.T., Calamari, D., Kaba, N., Mbome, I.L., Naeve, H., Ochumba, P.B.O., Osibanjo, O., Radegonde, V., Saad, M.A.H., 1994. Review of heavy metals in the African aquatic environment. Ecotoxicol. Environ. Saf. 28 (2), 134–159. https://doi.org/10.1006/eesa.1994.1041.

Cammilleri, G., Vazzana, M., Arizza, V., Giunta, F., Vella, A., Lo Dico, G., Giaccone, V., Giofrè, S.V., Giangrosso, G., Cicero, N., Ferrantelli, V., 2018. Mercury in fish products: what’s the best for consumers between bluefin tuna and yellowfin tuna? Nat. Prod. Res. 32, 457–462. https://doi.org/10.1080/14786419.2017.1309538.

Cao, P., Fujimori, T., Juhasz, A., Takaoka, M., Oshita, K., 2020. Bioaccessibility and human health risk assessment of metal(loid)s in soil from an e-waste open burning site in Agbogbloshie, Accra, Ghana. Chemosphere 240, 124909.

Chen, C.Y., Chen, Y.T., Chen, K.S., Hsu, C.C., Liu, L.L., Chen, H.S., Chen, M.H., 2018. Arsenic and five metal concentrations in the muscle tissue of bigeye tuna (Thunnus obesus) in the Atlantic and Indian Oceans. Mar. Pollut. Bull. 129, 186–193.

Cheyns, K., Banza, C.L.N., Ngombe, L.K., Asosa, J.N., Haufroid, V., De Putter, T., Nawrot, T., Kimpanga, C.M., Numbi, O.L., Ilunga, B.K., Nemery, B., Smolders, E., 2014. Pathways of human exposure to cobalt in Katanga, a mining area of the D. R. Congo. Sci. Total Environ. 490, 313–321.

Collette, B., Acero, A., Amorim, A.F., Boustany, A., Canales Ramirez, C., Cardenas, G., Car- penter, K.E., Chang, S.-K., Chiang, W., de Oliveira Leite Jr., N., Di Natale, A., Die, D., Fox, W., Fredou, F.L., Graves, J., Viera Hazin, F.H., Hinton, M., Juan Jorda, M., Minte Vera, C., Miyabe, N., Montano Cruz, R., Nelson, R., Oxenford, H., Restrepo, V., Schaefer, K., Schratwieser, J., Serra, R., Sun, C., Teixeira Lessa, R.P., Pires Ferreira Travassos, P.E., Uozumi, Y. & Yanez, E. 2011. Thunnus obesus. The IUCN Red List of Threatened Species 2011: e.T21859A9329255. https://dx.doi.org/10.2305/IUCN.UK.2011-2.RLTS. T21859A9329255.en.

Dhaneesh, KV, Noushad, KM, Biju Kumar, A, Nobi, EP, Nabeel, AM, 2014. Heavy metal con- tent in yellowfin tuna Thunnus albacares (Bonnaterre) of Lakshadweep Sea. Carpathian J. Earth Environ. Sci. 9 (4), 55–59.

Di Bella, G., Potortì, A.G., Lo Turco, V., Bua, D., Licata, P., Cicero, N., Dugo, G., 2015. Trace elements in Thunnus thynnus from Mediterranean Sea and benefit-risk assessment for consumers. 2015. Food Addit. Contam.: Part B 8 (3), 175–181.

Ekeanyanwu, R.C., Nwokedi, C.L., Noah, U.T., 2015. Monitoring of metals in Tilapia nilotica tissues, bottom sediments and water from Nworie River and Oguta Lake in Imo State, Nigeria. Afr. J. Environ. Sci. Technol. 9 (8), 682–690.

Elenge, M.M., de Brouwer, C., 2011. Identification of hazards in the workplaces of artisanal mining in Katanga. Int. J. Occup. Med. Environ. Health 24 (1), 57–66.

Elumalai, V., Brindha, K., Lakshmanan, E., 2017. Human exposure risk assessment due to heavy metals in groundwater by Pollution Index and multivariate statistical methods: a case study from South Africa. Water 9, 234. https://doi.org/10.3390/w9040234.

Esdaile, L.J., Chalker, J.M., 2018. The mercury problem in artisanal and small-scale gold mining. Chem. Eur. J. 24, 1–13. https://doi.org/10.1002/chem.201704840.

Estoup, A., Largiader, C.R., Perrot, E., Chourrout, D., 1996. Rapid one-tube DNA extraction for reliable PCR detection of fish polymorphic markers and transgenes. Mol. Mar. Biol. Biotechnol. 5, 295–298.

European Commission, 2020. A New Circular Economy Action Plan. For a Cleaner and more Competitive Europe. vol. 2020. COM 98 final, available at. https://eur-lex.eu- ropa.eu/legal-content/EN/TXT/?qid=1583933814386&uri=COM:2020:98:FIN accessed May 2020.

Galimberti, C., Corti, I., Cressoni, M., Moretti, V.M., Menotta, S., Galli, U., Cambiaghi, D., 2016. Evaluation of mercury, cadmium and lead levels in fish and fishery products imported by air in North Italy from extra-European Union Countries. Food Control 60, 329–337.

García, M.Á., Núñez, R., Alonso, J., Melgar, M.J., 2016. Total mercury in fresh and processed tuna marketed in Galicia (NW Spain) in relation to dietary exposure. Environ. Sci. Pollut. Res. 23, 24960–24969. https://doi.org/10.1007/s11356-016-7634-9.

Geslin V, Garcia-Vazquez E. 2020. Metal contamination in aquatic species from Africa. Mendeley Data, v2 doi:10.17632/4bpc9s4c49.2.

Gbogbo, S, Otoo, SD, Huago, RQ, Asomaning, O, 2017. High levels of mercury in wetland resources from three river basins in Ghana: a concern for public health. Environ. Sci. Pollut. Res. 24 (6), 5619–5627.

Gonzalez, E.G., Beerli, P., Zardoya, R., 2008. Genetic structuring and migration patterns of Atlantic bigeye tuna, Thunnus obesus (Lowe, 1839). BMC Evol. Biol. 8, 252. https://doi. org/10.1186/1471-2148-8-252.

Gu, Y.-G., Gao, Y.-P., 2018. Bioaccessibilities and health implications of heavy metals in exposed-lawn soils from 28 urban parks in the megacity Guangzhou inferred from an in vitro physiologically-based extraction test. Ecotoxicol. Environ. Saf. 148, 747–753.

Gu, Y.-G., Gao, Y.-P., Lin, Q., 2016. Contamination, bioaccessibility and human health risk of heavy metals in exposed-lawn soils from 28 urban parks in southern China's larg- est city, Guangzhou. Appl. Geochem. 67, 52–58.

Gull, A., Dar, A.A., Sharma, M., 2018. Effects of heavy metals on the health of pregnant women and fetus: a review. Int. J. Theoretical Appl. Sci. 10 (1), 1–9.

Gulley, A.L., McCullough, E.A., Shedd, K.B., 2019. China's domestic and foreign influence in the global cobalt supply chain.

Resour. Policy 62, 317–323.

Hall, T.A., 1999. BioEdit: a user-friendly biological sequence alignment editor and analysis program for Windows 95/98/NT. Nucleic Acids Symp. Ser. 41, 95–98.

Hammer, Ø., Harper, D.A.T., Ryan, P.D., 2001. PAST: Paleontological statistics software package for education and data analysis. Palaeontol. Electron. 4 (1), 9pp. http:// palaeo-electronica.org/2001_1/past/issue1_01.htm.

Jackson, M., Woodford, D.J., Weyl, O.L.F., 2016. Linking key environmental stressors with the delivery of provisioning ecosystem services in the freshwaters of southern Africa. Geo: Geography and Environment 3 (3), e00026.

Kahle, D., Wickham, H., 2013. Ggmap: spatial visualization with ggplot2. The R Journal 5 (1), 144–161. http://journal.r-project.org/archive/2013-1/kahle-wickham.pdf.

Kalonda, D.M., Tshikongo, A.K., Kule-koto, F.K., Numbi, O.L., Busambwa, C.K., Kalonda, O.K., Bwalya, Y.K., Cansa, H.M., Otshudi, A.L., Kalala, Z.L., 2017. Impact of mining on water of the rivers Shinkolobwe, Lwisha and Kansonga in the province of Katanga (DRC). J. Med. Res. 3, 71–73.

Kambole, M.S., 2003. Managing the water quality of the Kafue River. Phys. Chem. Earth 28, 1105–1109.

Kojadinovic, J., Potier, M., Le Corre, M., Cosson, R., Bustamante, P., 2007. Bioaccumulation of trace elements in pelagic fish from the Western Indian Ocean. Environ. Pollut. 146 (2), 548–566 (ff10.1016/j.envpol.2006.07.015ff. f).

Kyere, V.N., Greve, K., Atiemo, S.M., 2016. Spatial assessment of soil contamination by heavy metals from informal electronic waste recycling in Agbogbloshie, Ghana. Envi- ron. Health Toxicol. 31, e2016006.

Kyere, V.N., Greve, K., Atiemo, S.M., Ephraim, J., 2017. Spatial assessment of potential eco- logical risk of heavy metals in soils from informal e-waste recycling in Ghana. Envi- ron. Health Toxicol. 32, e2017018.

Lamborg, C.H., Hammerschmidt, C.R., Bowman, K.L., Swarr, G.J., Munson, K.M., Ohnemus, D.C., Lam, P.J., Heimbürger, L.-E., Rijkenberg, M.J.A., Saito, M.A., 2014. A global ocean inventory of anthropogenic mercury based on water column measurements. Nature 512, 65–68.

Le Croizier, G., Schaal, G., Gallon, R., Fall, M., Le Grand, F., Munarona, J.-M., Rouget, M.-L., Machud, E., Le Loc'h, F., Laë, R., Tito De Morais, L., 2016. Trophic ecology influence on metal bioaccumulation in marine fish: inference from stable isotope and fatty acid analyses. Sci. Total Environ. 573, 83–95.

Le Croizier, G., Schaal, G., Point, D., Le Loc'h, F., Machu, E., Fall, M., Munaron, J.-M., Boyé, A., Walter, P., Laë, R., Tito De Morais, L., 2019. Stable isotope analyses revealed the influ- ence of foraging habitat on mercury accumulation in tropical coastal marine fish. Sci. Total Environ. 650, 2129–2140.

Licata, P., Drombetta, D., Cristani, M., Naccari, C., Martino, D., Calo, N., et al., 2005. Heavy metals in liver and muscle of bluefin tuna (Thunnus thynnus) caught in the Straits of Messina (Sicily, Italy). Environ. Monit. Assess. 107, 239–248.

Maury, O., Gascuel, D., Marsac, F., Fonteneau, A., de Rosa, A.-L., 2001. Hierarchical interpre- tation of nonlinear relationships linking yellowfin tuna (Thunnus albacares) distribu- tion to the environment in the Atlantic Ocean. Can. J. Fish. Aquat. Sci. 58 (3), 458–469.

Musa Obadia, P., Kayembe-Kitenge, T., Haufroid, V., Banza, C.L.N., Nemery, B., 2018. Pre- eclampsia and blood lead (and other metals) in Lubumbashi. DR Congo. Environ. Res. 167, 468–471.

Nansai, K., Kondo, Y., Giurco, D., Sussman, D., Nakajima, K., Kagawa, S., Takayanagi, W., Shigetomi, Y., Tohno, S., 2019. Nexus between economy-wide metal inputs and the deterioration of sustainable development goals. Resour. Conserv. Recycl. 149, 12–19.

Nicklisch, S.C.T., Bonito, L.T., Sandin, S., Handoum, A., 2017. Mercury levels of yellowfin tuna (Thunnus albacares) are associated with capture location. Environ. Pollut. 229, 87–93.

Nikolic, N., Morandeau, G., Hoarau, L., West, W., Aguirrazabalaga, H., Hoyle, S., Nicol, S.J., Bourjea, J., Puech, A., Farley, J.H., Williams, A.J., Fonteneau, A., 2017. Review of alba- core tuna, Thunnus alalunga, biology, fisheries and management. Rev. Fish Biol. Fish. 27, 775–810. https://doi.org/10.1007/s11160-016-9453-y.

Nuñez, R., García, M.A., Alonso, J., Melgar, M.J., 2018. Arsenic, cadmium and lead in fresh and processed tuna marketed in Galicia (NW Spain): risk assessment of dietary expo- sure. Sci. Total Environ. 627, 322–331.

Orisakwe, OE., 2014. Lead and cadmium in public health in Nigeria: physicians neglect and pitfall in patient management. N. Am. J. Med. Sci. 6 (2), 61–70.

Ouali, N., Belabed, B.E., Zeghdoudi, F., Rachedi, M., 2018. Assessment of metallic contam- ination in sediment and mullet fish

(Mugil cephalus Linnaeus, 1758) tissues from the east Algerian coast. Journal of Water and Land Dev. 38 (1), 115–126. https://doi.org/ 10.2478/jwld-2018-0048.

Owusu, G., Donkor, A., Ziwu, C., Nyarko, S., Tawiah, R., Bonzongo, J.-C., Dubey, B., Asante, I., Nyame, F., 2017. Public perception of e-waste management and disposal practices in Accra metropolis, Ghana. Sci. Dev. 1, 79–87.

R Core Team, 2018. R: A Language and Environment for Statistical Computing. R Founda- tion for Statistical Computing, Vienna, Austria URL. https://www.R-project.org/. Roberts, S., Gunn, G., 2014. Cobalt. In: Gunn, G. (Ed.), Critical Metals Handbook. British Geological Survey, Wiley, pp. 122–149.

Saha, N., Zaman, M.R., 2013. Evaluation of possible health risks of heavy metals by consumption of foodstuffs available in the central market of Rajshahi City, Bangladesh. Environ. Monit. Assess. 185 (5), 3867–3878.

Schlüter, 2006. Geological Atlas of Africa, with Notes on Stratigraphy, Tectonics, Economic Geology, Geohazards and Geosites of each Country. Springer, Berlin, Heidelberg, New York (272pp).

Shütte, P., 2019. International mineral trade on the background of due diligence regula- tion: a case study of tantalum and tin supply chains from East and Central Africa. Resour. Policy 62, 674–689.

Sika, A.E., Ake Assi, Y., Koffi, K.M., Koffi-Nevry, R., Biego, H., 2014. Estimation of some heavy metals intake through tuna loins (Thunnus sp) produced in Côte D'ivoire. Int. J. Appl. Sci. Technol. 4 (3), 73–80.

Squadrone, S., Burioli, E., Monaco, G., Koya, M.K., Prearo, M., Gennero, S., Dominici, A., Abete, M.C., 2016a. Human exposure to metals due to consumption of fish from an artificial lake basin close to an active mining area in Katanga (D.R. Congo). Sci. Total Environ. 568, 679–684.

Squadrone, S., Brizio, P., Stella, C., Prearo, M., Pastorino, P., Serracca, L., Ercolini, C., Abete, M.C., 2016b. Presence of trace metals in aquaculture marine ecosystems of the north- western Mediterranean Sea (Italy). Environ. Pollut. 215, 73–83.

Sracek, O., et al., 2012. Mining-related contamination of surface water and sediments of the Kafue River drainage system in the Copperbelt district, Zambia: an example of a high neutralization capacity system. J. Geochem. Explor. 112, 174–188.

Srichandan, S., Panigrahy, R.C., Baliarsingh, S.K., Rao, B.S., Pati, P., Sahu, B.K., Sahu, K.C., 2016. Distribution of trace metals in surface seawater and zooplankton of the Bay of Bengal, off Rushikulya estuary, East Coast of India. Mar. Pollut. Bull. 111, 468–475.

Torres, P., Rodrigues, A., Soares, L., et al., 2016. Metal concentrations in two commercial tuna species from an active volcanic region in the mid-Atlantic Ocean. Arch. Environ. Contam. Toxicol. 70, 341–347.

Viñas, J., Tudela, S., 2009. A validated methodology for genetic identification of tuna spe- cies (genus Thunnus). PlosOne 4 (10), e7606. https://doi.org/10.1371/journal. pone.0007606.

Von Stackelberg, K., Li, M., Sunderland, E.M., 2017. Results of a national survey of high- frequency fish consumers in the United States. Environ. Res. 158, 126–136.

Wang, X., Sato, T., Xing, B., Tao, S., 2005. Health risks of heavy metals to the general public in Tianjin, China via consumption of vegetables and fish. Sci. Total Environ. 350 (1–3), 28–37.

Wang, X.-N., Gu, Y.-G., Wang, Z.-H., 2020. Fingerprint characteristics and health risks of trace metals in market fish species from a large aquaculture producer in a typical arid province in Northwestern China. Environ. Technol. Innov. 19, 100987.

World Health Organization, Food and Agriculture Organization of the United Nations, Joint FAO/WHO Expert Committee on Food Additives, 2011. Evaluation of certain food additives and contaminants: seventy-second [72nd] and seventy- rd [73rd] re- ports of the Joint FAO/WHO Expert Committee on Food Additives. World Health Or- ganization https://apps.who.int/food-additives-contaminants-jecfa-database/ chemical.aspx?chemID=1806 https://apps.who.int/iris/handle/10665/44515 accessed June 2018.

Zagaglia, C.R., Lorenzzetti, J.A., Stech, J.L., 2004. Remote sensing data and longline catches of yellowfin tuna (Thunnus albacares) in the equatorial Atlantic. Remote Sens. Envi- ron. 93 (1–2), 267–281. https://doi.org/10.1016/j.rse.2004.07.015.

Zeuner, B., 2018. An obsolescing bargain in a rentier state: multinationals, artisanal miners, and cobalt in the Democratic Republic of Congo. Front. Energy Res. 6, 123. https://doi.org/10.3389/fenrg.2018.00123.

Zohra, BS, Habib, A, 2016. Assessment of heavy metal contamination levels and toxicity in sediments and fishes from the Mediterranean Sea (southern coast of Sfax, Tunisia). Environ. Sci. Pollut. Res. Int. 23 (14), 13954–13963.

Table 1

Tuna samples analysed in this work (FAO fishing areas: see 1.2). Thunnus alalunga: longfin or albacore tuna; T. albacares: yellowfin tuna; T. obesus: bigeye tuna.

Waters	FAO fishing area	Location	Species	Sample size
Europe	27	North of the Azores (subarea XII)	*Thunnus alalunga*	14
Africa	34	Morocco/Mauritania	*T. albacares*	13
			T. obesus	3
	47	Gulf of Guinea	*T. alalunga*	3
Oceania	77	Cook Islands	*T. albacares*	19

Table 2

Results of accuracy assessment (see 2.4).

	Cu	Zn	Ag	Cd	Hg
UΔ	0.3241	2.2027	1.787	0.0038	0.0796
Δm	0.17	2.18	0.33	0.002	0.17

Table 3

Metal contents in muscle tissue of tuna species caught by Spanish vessels in different FAO areas (see 1.2; Table 1) and sold in Spanish markets. Data are given in μg/kg of wet weight (CV % in parenthesis). In italics, Hg values corrected according to the accuracy assessment dividing uncorrected values by 1.28.

Species	*Thunnus albacares*		*Thunnus alalunga*		*Thunnus obesus*
Capture zone	FAO 34	FAO 77	FAO 27	FAO 47	FAO 34
As	469.69 (3.71)	667.98 (3.43)	1501.15 (1.63)	2443.31 (2)	696.97 (2.8)
Cd	10.57 (9.6)	9.93 (8.1)	22.01 (5.28)	7.73 (9.2)	46.49 (7.57)
Co	2.83 (12.7)	3.71 (12.03)	7.92 (9.7)	3.57 (11.4)	1.96 (12.26)
Cr	43.44 (7.2)	91.02 (5.24)	31.74 (4.6)	25.55 (8.6)	45.9 (7.6)
Cu	345.39 (3.87)	365.88 (3.76)	559.54 (1.5)	386.75 (4.3)	283.42 (2.57)
Ni	54.7 (5.3)	71 (7.9)	459.35 (5.1)	20.39 (12.2)	42.99 (10.37)
Hg	644.79 (4.38)	447.4 (3.4)	214.41 (2.48)	462.86 (0.4)	1157.54 (2.87)
	503.7 (4.55)	*349.5 (4.71)*	*167.5 (1.38)*	*361.6 (0.34)*	*904.3 (2.12)*
Pb	8.79 (9.35)	6.55 (8.9)	4.09 (8.73)	10.29 (3.6)	1.47 (18.4)
Zn	5012.3 (3.5)	4793.94 (3.04)	8292.4 (1.56)	6671.3 (3)	5512.2 (1.8)

Table 4

Two-factor ANOVA comparing T. alalunga and T. albacares samples from African and non- African waters. Results are presented by metal. * p < .05, ** p < .01, *** p < .001, ns = not significant.

	Species		Continent		Interaction	
Metal	SSQ	F	SSQ	F	SSQ	F
As	12,824,800	138.1 ***	346,178	3.7 ns	3,162,860	34.1 ***
Cd	969.5	1.9 ns	291.9	0.6 ns	486.4	0.97 ns
Co	70.01	2.04 ns	97.3	2.8 ns	11.7	0.3 ns
Cr	19,063.4	8.9 **	6856.4	3.2 ns	4625.3	2.2 ns
Cu	326,264	21.1 ***	96,609.8	6.2 *	62,922.5	4.05 ns
Hg	797,220	19.3 ***	737,647	17.9 ***	72,325.8	1.8 ns
Ni	1,215,620	1.9 ns	370,053	0.6 ns	369,074	0.6 ns
Pb	58.5	2.7 ns	133.9	6.1 *	35.8	1.6 ns
Zn	108,250,000	16.8 ***	9,827,720	1.5 ns	12,249,200	1.9 ns

Table 5

Maximum Tolerable Weekly Intake, in kg of tuna (Thunnus spp.) from different capture zones analysed in this study, for an adult weighing 70 kg (see text). Estimates are presented for the three metals considered in EU regulation EC No 1881/2006. Hg_u and Hg_c, uncorrected and corrected Hg contents respectively.

	Cd	Hg_u	Hg_c	Pb
T. albacares FAO 34	36.073	0.551	0.705	324.165
T. albacares FAO 77	55.397	0.823	1.053	303.474
T. alalunga FAO 27	43.802	1.623	2.077	482.238
T. alalunga FAO 47	53.708	0.769	0.984	170.087
T. obesus FAO 34	8.774	0.276	0.353	1186.663

Fig. 1. A: Maps of metals (As, Cd, Co above; Cu, Pb, Hg below) found in African Actinopterygii from 1990 to 2017 (for references, see Geslin & García-Vázquez 2020). B: Map showing mining activities in Africa (different goods are shown as different colours; source U.S. Geological Survey Open-File Report 2006–1135, https://pubs.usgs.gov/of/2006/1135/). Circle diameters are proportional to metal concentration (A) or extraction capacity (B).

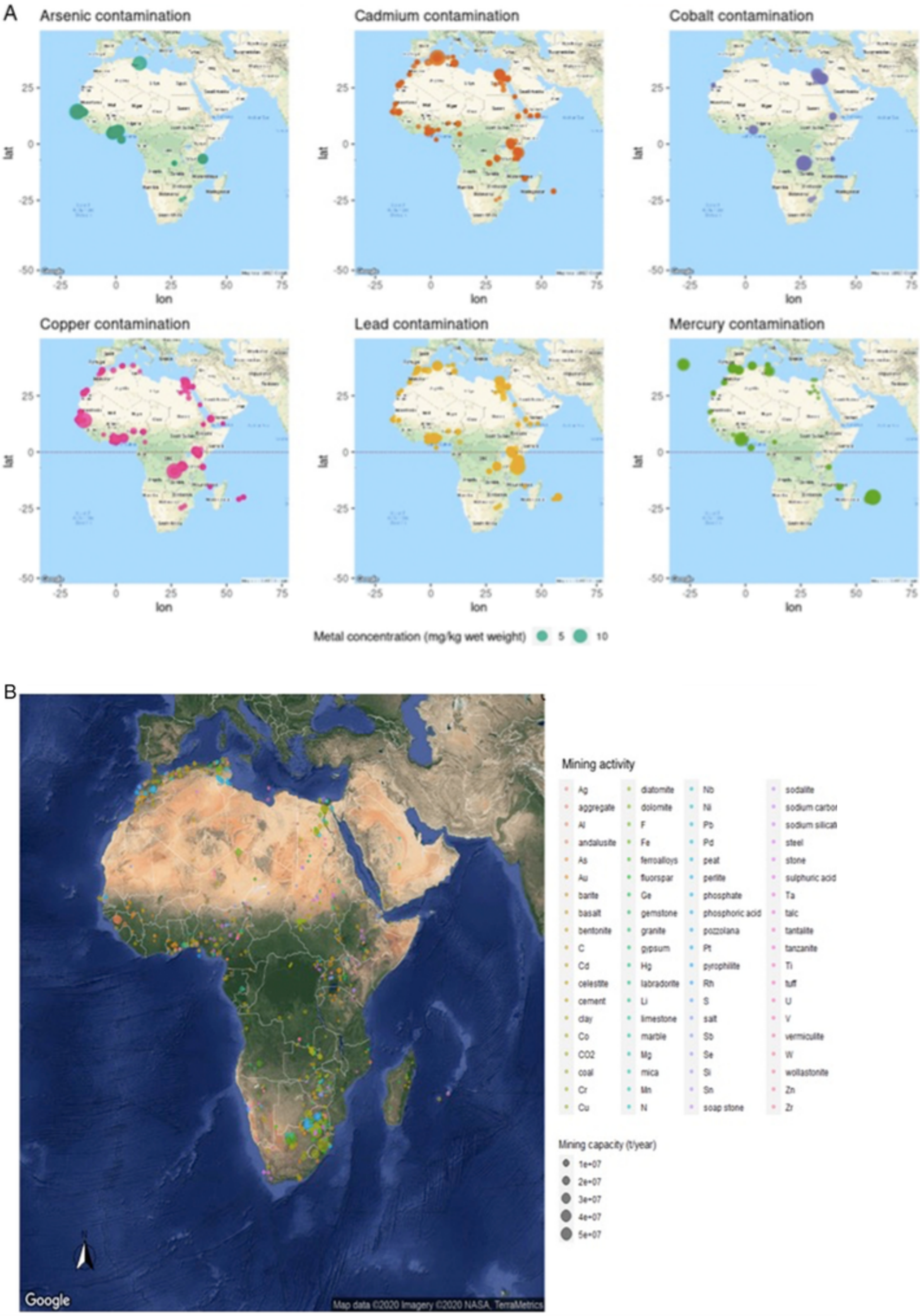


Fig. 2. Metadata analysis of published fish metal contents: A) evolution of the number of analysed samples over the decades, as the proportion of samples from each decade for each pollution source ("diffuse" includes agricultural, urban and background pollution); B) average metal content per main pollution source (5% error as capped bars); please note logarithmic scale. Sample sizes range from 1 to 50 fish.

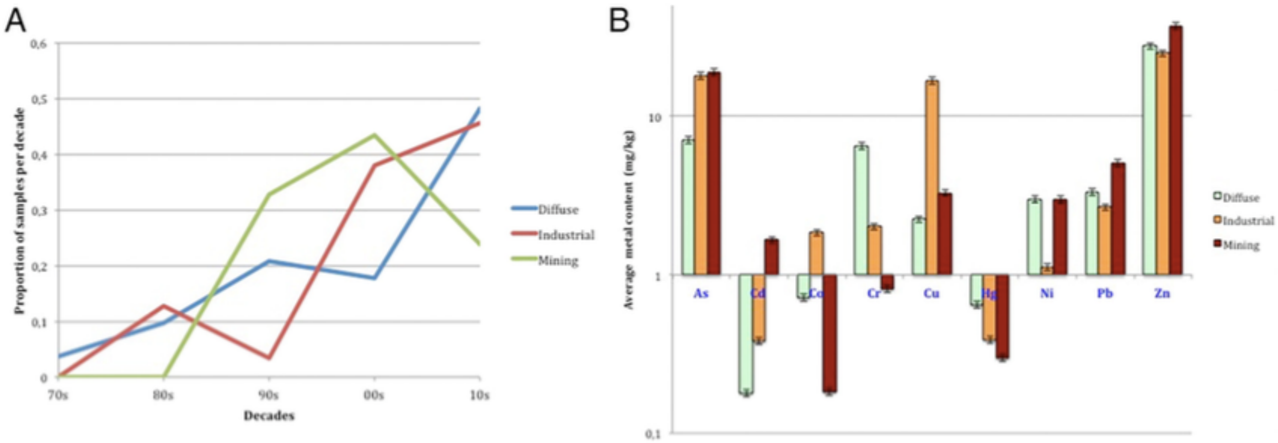


Fig. 3. Scatter plot of PC1 on PC2 obtained from As, Cd, Co, Cu, Hg and Pb contents in muscle tissue of tuna sold in Spain. Squares: Thunnus alalunga (blue: FAO fishing area 27, beige: FAO 47); dots: T. albacares (brown: FAO 77, black: FAO 34); diamonds: T. obesus. Diagonals represent the relative contribution of each metal to the observed variance.

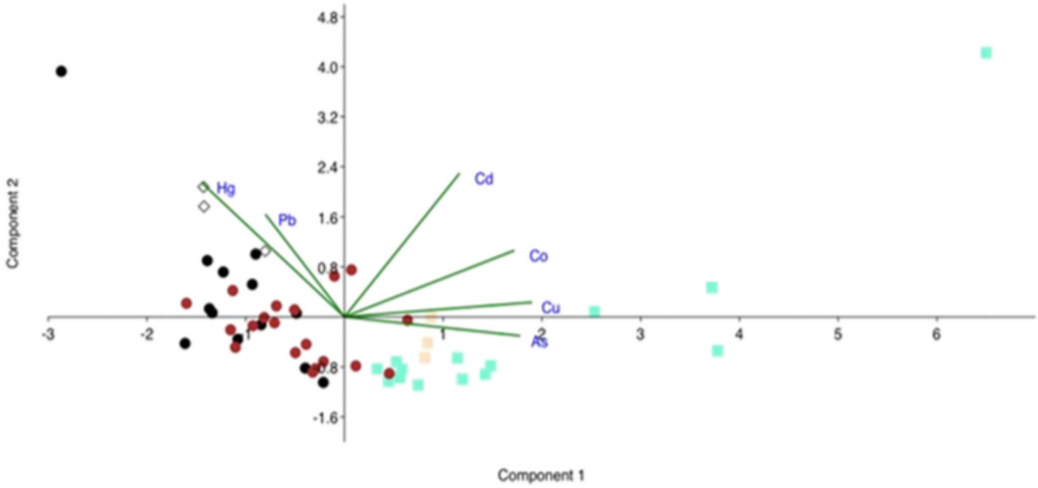
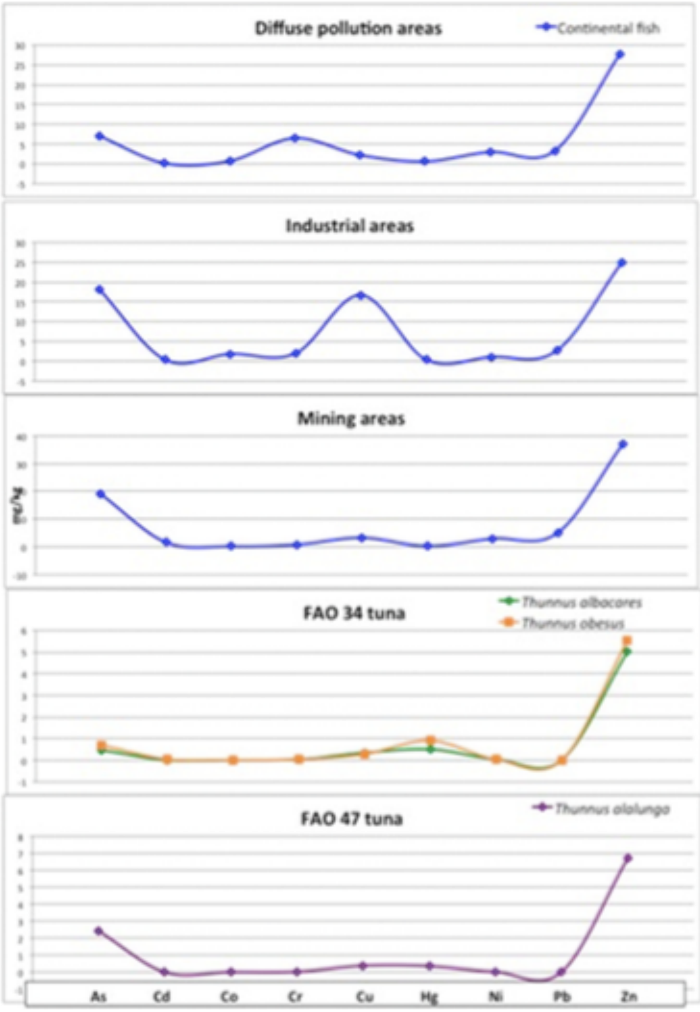


Fig. 4. Trace metal fingerprints of African continental fish inhabiting waters with diffuse pollution (top), industrial pollution (second from the top) and mining pollution (third from the top); tuna samples from FAO 34 area in front of African coasts (second from the bottom) and from FAO 47 area in the Gulf of Guinea (bottom). Metal contents are expressed as mean mg/kg. Metadata for the continental fish can be found in Geslin and Garcia-Vazquez (2020); tuna data are from this study.



Diffuse pollution areas
Continental fish
Industrial areas
Mining areas
mg/kg
FAO 34 tuna
Thunnus albacares
Thunnus obesus
FAO 47 tuna
Thunnus alalunga
As
Cd
Co
Cr
Cu
Hg
Ni
Pb
Zn